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LIGHT-MATTER SEMINAR

Title: New Experimental Platforms for Molecular Polaritonics

Abstract: Polaritons are hybrid light-matter states with unusual properties that arise from strong interactions between a molecular ensemble and the confined electromagnetic field of an optical cavity. Cavity-coupled molecules appear to demonstrate energetics, reactivity, and photophysics dramatically distinct from their free-space counterparts, but the mechanisms and scope of these phenomena remain open questions. I will discuss new experimental platforms that the Weichman Lab is developing to investigate molecular reaction dynamics under vibrational strong coupling.

While polaritons are now well-established in solution-phase and solid-state systems, they had not been previously reported in isolated gas-phase molecules, where attaining sufficiently strong light-matter interactions is a challenge. We access the strong coupling regime in an intracavity cryogenic buffer gas cell optimized for the preparation of simultaneously cold and dense ensembles and report a proof-of-principle demonstration in gas-phase methane. We strongly cavity-couple individual rovibrational transitions and probe a range of coupling strengths and detunings. In ongoing work, we will harness this infrastructure as a new testbed for fundamental studies of polariton physics and chemistry.

We are also searching for signatures of cavity-altered dynamics in benchmark solution-phase systems. So far, we have focused on radical hydrogen-abstraction processes, which have well-characterized reactive surfaces and can be initiated with photolysis and tracked directly on ultrafast timescales. We use ultrafast transient absorption to examine intracavity reaction rates with the goal of better understanding exactly how and when reactive trajectories may be influenced by strong light-matter interactions.